Research Article

Rapid Electrochemical Detection of Radiolysis Products in an Aqueous Solution Exposed to Alpha Particle Beams

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An electrochemical cell has been developed that allows for the rapid and exhaustive detection of oxygen and/or hydrogen peroxide produced during the radiolysis of aqueous solutions by alpha particle or proton beams. Short, 10–100 s, exposures were sufficient to yield steady-state electrochemical currents proportional to the radiolysis G-factor. The use of thin SiC windows provided a robust means of separating the vacuum environment of the ion-beam accelerator from the aqueous solution at atmospheric pressure with minimal energy loss.

1. Introduction

Radiolysis of water, the production of hydrogen and oxygen from the splitting of water by the decay products of radioactive materials, has primarily been studied from the point of view of radioactive waste management [1]. Water is often in contact with radioactive waste, and the obvious hazards associated with potentially explosive (from H2/O2 mixtures) or corrosive (H2O2 is often also produced in the radiolysis process) situations required study of the radiolysis process. Concurrently, the possibilities for the commercial production of hydrogen via radiolysis have been considered [2]. In this scenario, electrical energy production is the end goal, with the hydrogen produced by radiolysis being intended as fuel for an electrochemical fuel cell.

Utilizing radioisotopes to generate electrochemical cell reactants is not a new concept. Extensive research has been conducted in radiation chemistry, analyzing the production of chemical species through radiolytic interactions [3–5]. During the 1950s and 1960s, research was conducted utilizing radiochemical means to power fuel cells. These systems were designed to utilize radioactive waste to produce power on a large scale. Not only was the water cycle studied, as is investigated here, but so were the ferric ion and ozone cycles [3]. In these systems, chemical intermediates produced via radiolysis were separated and fed to fuel cells in a normal fashion. The overall efficiency of these units was <1% and would require significant improvements to become a cost effective means of large-scale power generation. Recently however, advances in microfabrication have been employed to take a fresh look at power generation via radiolysis. Using these same radiolysis principles in conjunction with microfabrication, it may be possible to develop a micropower source capable of continuous power output for several years [6, 7]. The idea is to convert nuclear energy into electrical energy via a chemical intermediate. Exposure to radiation results in the radiolytic decomposition of water, producing chemical intermediates H2, O2, and H2O2 [4]. The H2, O2, and H2O2 can then be used to power a mixed reactant fuel cell. The system is regenerative since the H2, O2, and H2O2 produced by radiolysis are converted back to water by the fuel cell reaction, closing the cycle. The regenerative cell is then capable of producing electricity for the life of the radioisotope. This closed cycle is illustrated in Figure 1.

The efficiency of chemical production via radiolysis can differ greatly from one type of radiation to another. This is especially apparent when comparing radiation with high
linear energy transfer (LET), such as alpha (He\(^{++}\) particles), and those with low LET, such as beta and gamma. Low LET radiation interacts with water molecules in isolated events that can be far from one another. A majority of the ions and radicals created during these separate events then diffuse into the bulk where they can react, resulting in low chemical yields. In the case of high LET radiation, interactions with water molecules are more constant, and large channels of ions and radicals (spurs) are created. The ions and radicals then react with one another within the spur and very little diffuses into the bulk, resulting in higher chemical yields. The exact mechanism by which molecular species are formed is still under some debate [8]. However, an overview of molecular production, including some of those mechanisms proposed in the literature, can be useful. The process starts with the production of H and OH radicals either by direct cleavage of the H-OH bond or loss of an electron and subsequent decomposition (2)–(4):

\[
\begin{align*}
H_2O &\rightarrow H + OH, \\
H_2O &\rightarrow H_2O^+ + e^-, \\
H_2O^+ + H_2O &\rightarrow OH + H_3O^+, \\
e^- + H_2O &\rightarrow H + OH^-.
\end{align*}
\]

(1) (2) (3) (4)

The H and OH radicals then combine to form molecular products:

\[
\begin{align*}
H + H &\rightarrow H_2, \\
OH + OH &\rightarrow H_2O_2, \\
H + OH &\rightarrow H_2O.
\end{align*}
\]

(5)

The production of O\(_2\) is also widely reported in the literature, but it is not clear whether O\(_2\) is produced directly from radical combination, or through the decomposition of H\(_2\)O\(_2\). Some theory suggests possible formation of O radicals which could combine to form O\(_2\), but this is still under discussion [4, 8].

The chemical yield is generally reported as a \(G\) factor, or the number of molecules formed per 100 eV of energy deposited. In the case of pure water, if \(G(H_2) = 1\), then \(\approx 2.5\%\) of the energy deposited was used to split water and produce H\(_2\) and H\(_2\)O\(_2\)/O\(_2\). Literature tells us that \(G(H_2) < 1.7\) for radiolysis of pure water by any form of radiation [4, 5]. The other products simply recombine to form water. In the case of alpha emission, the generation of H\(_2\) is about 6–10x that of O\(_2\) [9]. The balance is then hydrogen peroxide. In the case of 6x production of H\(_2\), the net reaction is

\[
10H_2O \xrightarrow{\alpha} 6H_2 + O_2 + 4H_2O_2.
\]

(6)

In literature studies of alpha radiolysis yield, typically samples are enclosed in a glass vial and irradiated. The irradiated vial was then crushed [1, 10] or extracted \(via\) syringe [2, 11] and its contents injected into a gas chromatograph or mass spectrometer. Using this method, throughput of samples is relatively slow, and there is the possibility of significant error. Thus, in order to measure radiolysis yield, a novel experimental method has been developed. The experimental cell developed for this study allows for rapid evaluation of radiolysis yield and considerably higher throughput of samples.

2. Experimental

The cell shown in Figure 2 was coupled to one of the beam lines on the NEC SSDH accelerator in the Department of Materials Science and Engineering at CWRU. The cell is fabricated in the form of a 2.75\” Conflat flange. Since the electrochemical cell has to be nonconductive and stable to chemical attack by the electrolytes used, the blank flange was machined out of a solid block of chlorinated-polyvinyl chloride (CPVC). Machined into the same block as the electrochemical cell were recesses for a quartz window to visualize the ion beam spot and a Faraday cup to measure beam current. These were sealed into the block using high vacuum epoxy.

The NEC accelerator is capable of producing an alpha particle beam up to 5.1 MeV and with beam currents of up to 100 nA. Proton beams of up to 3.4 MeV can also be
Figure 2: (a) Electrochemical cell for radiolysis measurements. A: 2.75” Conflat blank flange, machined from CPVC block. B: electrolyte channel—1 cm long, 2 mm wide, 2 mm deep. C: recess for Faraday cup. D: recess for quartz window. (b) Detailed view of Electrochemical cell for radiolysis measurements. Drawing not to scale.

generated. The focused alpha beam had a typical diameter of 1 mm and a current of 1–5 nA. The ion beam current measured by the Faraday cup corresponds to 2e− per He++ or 1e− per H+. The beam current was typically measured before and after a series of four radiolysis exposures. A beam current measurement consisted of the average value of thirty current measurements made at a 1 Hz sampling rate.

The radiolysis solution is separated from the accelerator vacuum by a 3 μm thick film of silicon carbide, (SiC) with an unsupported area of 2.5 mm × 2.5 mm square centered in a 1 cm × 1 cm Si chip as shown in Figure 2(b). To prepare these windows, a film of SiC is grown by an atmospheric pressure CVD process on one face of a silicon wafer. On the back side of the wafer, a mask is applied, and the Si substrate is removed using a KOH etch to expose a region of the SiC film [12]. The SiC film is completely impervious to the KOH etch. For the SiC unsupported area and thickness used in these experiments, a pressure differential across the SiC film of >207 kPa was found to be necessary to break the SiC film. This provided a generous 2:1 safety margin when operating the electrochemical cell at atmospheric pressure. The Si chip with the SiC window was mounted to the cell with vacuum epoxy. The cell is mounted on an X-Y positioning stage on the end of the beam line that allows the beam to be centered on the SiC window, or on the quartz window or the Faraday cup. A shutter within the beam line is used to control the exposure time when the electrochemical cell was irradiated. On the solution side of the SiC film were a 3 nm thick Ti adhesion layer and a 100 nm thick silver film that was used as the working electrode. Both of these films were applied by vacuum deposition. When the Si/SiC chip was affixed to the cell, an electrical connection to the working electrode (Ag film) was made with silver epoxy to a Ag wire imbedded in the CPVC block.

After passing though the thin SiC, Ti, and Ag films, the alpha particles enter the aqueous solution, and radiolysis occurs close to the working electrode. In this arrangement, O2 and/or H2O2 that are radiolytically generated are detected by being electrochemically reduced at the silver film. In alkaline solutions, the reference electrode was a silver wire on which a thin Ag2O film had been grown. As the silver working electrode has a wide potential range (≈0 V to 0.7 V versus a reversible hydrogen electrode (RHE); see Figure 3) over which O2 and H2O2 reduction is the only reaction that occurs, exacting potential control is not required, and the Ag/Ag2O reference electrode provides a sufficiently stable potential of ≈1.15 V versus RHE. For measurements in hydrochloric acid solutions, a thin film of AgCl was electrochemically formed on the Ag wire, creating a reference electrode with a stable potential based on the Ag/AgCl/Cl− couple. The counter electrode was a Pt wire ≈1 cm from the working electrode which minimized any disturbances resulting from the counter electrode reaction. All solutions were deaerated with bubbling argon for at least 30 minutes prior to being pumped into cell. During the radiolysis experiments, the solution was not circulated, but was stagnant within the cell. All electrochemical measurements were made with a Solartron 1280B potentiostat.
3. Results and Analysis

3.1. Verification of the Electrochemical Technique. To demonstrate that silver is electrochemically active for the reduction of oxygen and hydrogen peroxide, but not for the oxidation of hydrogen, cyclic voltammetry was conducted over a range of 0.0–1.4 V (versus RHE) at 50 mV/s in solutions of varying pH that were saturated with nitrogen (base case), oxygen, or hydrogen, or that had 1 or 10 mM H$_2$O$_2$ added. Typical results are illustrated in Figures 3(a) and 3(b).

At potentials less than 0.8 V versus RHE, Ag exhibits excellent O$_2$ reduction, on par with Pt. In addition, Ag shows essentially no activity for H$_2$ oxidation. Ag is also an excellent electrocatalyst for H$_2$O$_2$ reduction and results in similar electrochemistry to that obtained for the oxygen saturated solution. Since Ag and Ag$_2$O are not electrochemically active for H$_2$ oxidation as shown, the H$_2$ that is radiolytically generated simply remains in the electrolyte. In alkaline solutions, the Ag/Ag$_2$O couple is ≈ 1.15 V versus RHE. As a result, for radiolysis experiments in alkaline solutions, the potential of the silver electrode was held at −1.0 V versus Ag/Ag$_2$O equivalent to +0.15 V versus RHE for reference to Figure 3.

For the radiolysis experiments conducted in this study, a 4.5 MeV alpha beam was used. After traveling through the SiC window, the Ti film (negligible energy loss), and the Ag film, the incident energy on the electrolyte was 3.7 MeV. With an incident energy of 3.7 MeV, alpha particles travel $\sim$ 25 μm through water, losing their energy in a nearly linear fashion. Radiation penetration depths and energy losses were calculated using stopping powers provided by The National Institute of Standards and Technology (NIST) [13].

The current observed during beam exposure is on the order of μA and is due to electrochemical reaction and not due to the beam itself (the beam current is on the order of 1 nA). As is illustrated by Figure 4, a plot of reduction current versus beam current is linear through the origin with little variation. If the beam itself had some direct impact on the electrochemical current, one would expect a nonlinear response with respect to beam current, which is not the case.

This experimental setup and method allow for rapid detection of radiolysis products at the working electrode and for minimization of losses due to diffusion. O$_2$ and H$_2$O$_2$ are generated within 25 μm of the working electrode, which is operated at the limiting current. This proximity allows for efficient capture of essentially all of the radiolytically generated O$_2$ and/or H$_2$O$_2$. The efficient collection of O$_2$/H$_2$O$_2$ can be proved in several ways. First, transient, one-dimensional concentration profiles within the cell during operation were simulated based on Fick’s second law with generation:

$$\frac{\partial C_A}{\partial t} - D_A \frac{\partial^2 C_A}{\partial x^2} = R_g,$$  \hfill (7)
Collection efficiency (%)

Exposure time (seconds)

Figure 5: Simulated collection efficiency of the radiolytic yield measurement cell with respect to time exposed to the alpha particle beam; Solid: H₂O₂, dashed: O₂.

where \( R_g \) [mol cm\(^{-3} \) s\(^{-1} \)] is the volumetric generation of component \( A \), \( D_A \) [cm\(^2\) s\(^{-1} \)] is the diffusivity of component \( A \), \( t \) [sec] is time, and \( C_A \) [mol/cm\(^3\)] is the concentration of component \( A \). Details of the simulation procedure can be found in [7]. Results of such a simulation give an estimation of the system's collection efficiency with respect to exposure time as illustrated in Figure 5.

Generation of O₂ and H₂O₂ occurs nearly uniformly in the 25 μm nearest to the working electrode (the radiolysis zone). The working electrode always operates at limiting current, that is, the concentration at the electrode surface is zero, so there is a competition between diffusion toward the working electrode and diffusion into the bulk liquid, where the concentration is also zero. Initially, the concentration of these products is very low, and although the working electrode is operating at limiting current, the driving force for diffusion toward the working electrode is relatively low, and only a small percentage of the O₂ and H₂O₂ generated is collected by the working electrode. As exposure to the alpha beam continues, the concentration of O₂ and H₂O₂ quickly rises in the radiolysis zone, with peak concentration occurring at the edge of the radiolysis zone. As the working electrode continues to operate at limiting current, the concentration gradient grows, increasing the driving force for diffusion towards the working electrode, which increases the percentage of O₂ and H₂O₂ collected by the working electrode. This process occurs rapidly, and high collection efficiencies are quickly established. In our simulations, after just three seconds of exposure, collection efficiency is already >87%, after ten seconds of exposure, collection efficiency is >93%, and at longer times, the efficiency is >95%. Evolution of the H₂O₂ concentration profile over the first thirty seconds of alpha beam exposure is illustrated in Figure 6. Evolution of the O₂ concentration profile is qualitatively similar.

High collection efficiency can also be observed in the experimental results. The result of a 58-second exposure of 0.01 M KOH to a 1.28 nA alpha beam is shown in Figure 7. When the shutter was opened at approximately 8.5 seconds, the response to the exposure was nearly instantaneous and steady-state was quickly achieved, in good agreement with the simulations. The small variations in the electrochemical current at steady-state were due to variations in the alpha beam current. When the shutter was closed, the electrochemical response quickly returned to the baseline. The small “tail” after exposure resulted from the collection of a small amount of radiolysis product that had diffused a short distance away from the working electrode. The fast onset of steady-state and insignificant tail after exposure imply very high collection efficiency.

Further experimental proof of high collection efficiency is presented when comparing the tail after exposures of different duration as shown in Figure 8. Since the ion beam current varied over the course of these measurements, the results are presented with the electrochemical current normalized to the ion beam current.
There is no significant difference between the diffusion tails illustrated in Figure 8. If the collection efficiency were not extremely high, there would be an accumulation of O$_2$ and H$_2$O$_2$ away from the electrode that would increase with increasing exposure time to the alpha beam. This would result in a tail that would increase in size with increasing exposure duration. However, there is no appreciable increase in the diffusion tail size with increasing exposure time for this system, confirming that collection efficiency of O$_2$ and H$_2$O$_2$ is extremely high.

It should be noted that if the penetration depth were much greater than 25 µm, the collection efficiency would be lower and a more pronounced diffusion tail would be seen. For example, consider the electrochemical responses to a 3.0 MeV proton beam versus a 1.2 MeV proton beam as shown in Figure 9. The 3.0 MeV proton beam would have a penetration depth of ~26 µm, similar to that of 3.7 MeV alpha particles. From Figure 9, it is clear that a significant diffusion tail is present when the radiation penetration depth is large, indicating low collection efficiency by the working electrode. On the other hand, when the penetration depth is smaller (~26 µm), the response shows very little diffusion tail, indicating very high collection efficiency.

Since the collection efficiency of O$_2$/H$_2$O$_2$ is very high, the steady-state current achieved during exposure to the alpha beam translates directly to the radiolysis yield of O$_2$ and H$_2$O$_2$. The balance of the O$_2$/H$_2$O$_2$ reaction is H$_2$ as given by (7), making the calculation of G(H$_2$) trivial. It should be noted that the relative yield of O$_2$ or H$_2$O$_2$ is immaterial to the calculation of the hydrogen yield, as one H$_2$ molecule was produced for every two electrons detected in the electrochemical current, regardless of whether O$_2$ or H$_2$O$_2$ was reduced. In addition, two electrons are required to neutralize one He$^{++}$ in the measurement of the beam current, so that the G-factor can be calculated using the following equation assuming perfect collection efficiency:

$$G(H_2) = 100 \times \left( \frac{\text{Steady-state Electrochemical current}}{\text{Incident Beam Energy} \times \text{Beam Current}} \right),$$

where $G$ is in molecules H$_2$/100 eV deposited energy and the electrochemical current has been corrected for the small background current present before radiolysis.

3.2. Radiolysis Yield of Solutions of KOH, NaOH, and HCl. In order to examine the effect of pH on the behavior of the electrochemical cell and the radiolysis yield, experiments were conducted to measure the yield for solutions of HCl, KOH or NaOH of various ionic strength. The error calculated for all experimental data is based on a 95% confidence interval:

$$C_{I0.95} = \pm 1.96\sigma,$$

where $C_{I0.95}$ is the 95% confidence interval and $\sigma$ is the standard deviation. Typically, 20 to 30 separate exposures were
used to generate the average yield and standard deviation. It is important to note that the standard deviation and not the standard error was used to calculate the confidence interval. This means that the confidence interval applies for individual measurements and not for averages.

Solutions of pH = 11, 12, 13, and 14 (0.001, 0.01, 0.1, and 1 M) were prepared with NaOH and KOH. It was found that there was no statistical difference in the radiolysis yield between KOH and NaOH solutions of equal concentration. When exposed to the alpha beam, solutions of pH = 14 (1 M) exhibited a fairly sharp response and achieved steady-state in 8 seconds. This response was not as sharp as was observed with solutions of pH = 13 or 12. This is believed to be due to somewhat lower diffusivities of O2 and H2O2 in the more concentrated alkaline solution.

When exposed to the alpha beam, solutions of pH = 12 and 13 exhibited a sharp response and quickly (<=5 seconds) achieved steady-state.

Solutions of pH = 11 (1 mM) exhibited a somewhat sluggish response and achieved steady-state in 15-20 seconds. The sluggish response is due to the low conductivity of 1 mM KOH (or NaOH) solutions. In this system, the primary carriers of ionic current are the cation and OH-. At low concentrations, it takes some time for the ionic current carrying species to achieve appropriate concentration profiles to sustain higher currents, resulting in the sluggish response. However, once these concentration profiles are established, the steady-state current still corresponds with the generation rate of O2 and H2O2. The O2 and H2O2 that were able to diffuse away during the initial sluggish response are recaptured after exposure in the diffusion tail. For this solution, optimal results were obtained with relatively low beam currents (<=1 nA) that minimized the electrochemical current. This in turn minimized the voltage drop resulting from the flow of current between the working electrode and the counter electrode across the solution resistance between those electrodes. At higher beam currents, the electrochemical current and resulting voltage loss were sufficiently large that the 30 V compliance limit of the potentiostat was reached, and the working electrode potential could not be maintained at the desired value.

Solutions of pH = 2 (10 mM) were prepared with HCl. When exposed to the alpha beam, the electrochemical response from solutions of pH = 2 are very similar to those of pH = 12 (also 10 mM), and exhibited a sharp response and quickly achieved steady-state. This is not surprising since the ionic conductivities and oxygen diffusion coefficients are very similar for pH = 2 and pH = 12. For 0.01 M HCl solutions, the working electrode was potentiostated at ~0.4 V versus Ag/AgCl/Cl- which gave a similar overpotential for peroxide reduction as was employed in alkaline solutions.

From the results obtained with pH = 2, 11, 12, 13, and 14, the dependence of G(H2) on pH is shown in Figure 10. It is clear that increases in pH beyond 11 result in lower and lower radiolysis yield. The reason for this lower yield is not clear, nor is it the aim of this work to explain the mechanism. However, this effect may be due to greater radical scavenging by the ions present in solution.

The radiolysis yield appears to level off closer to neutral pH, and low total ion concentration is approached. The data indicates that for a pure water solution, the radiolysis yield would likely be $G(H_2) \sim 1$. Considering the high end of the 95% confidence intervals, this result is consistent with the lower end of reported literature values for pure water, which are shown in Figure 10 as the point at pH 7. Furthermore, the G factors reported here were calculated assuming 100% collection efficiency while the simulations suggest that, at steady-state, the true collection efficiency is slightly lower, on the order of 97%. Correcting for the collection efficiency would raise the calculated G-factor slightly closer to literature values.

### 4. Discussion

This new method of detecting radiolysis yield has many advantages over the method of irradiating vials described in the literature. With the previous method, relatively long exposures, on the order of hours, may be required in order to build up sufficient hydrogen for analysis by gas chromatograph. This method achieves rapid detection of radiolysis products that is both accurate and efficient. There is inherently a realtime evaluation of radiolysis yield. This could be useful if, for instance, the electrolyte contains scavenging species that are depleted during exposure. The method also provides a very accurate measure of beam stability and can easily see events such as the beam “winking” off and on, beam fluctuation, or beam decay. New electrolytes and solutions of interest can be easily pumped through the cell, allowing for rapid screening. It should be noted that solutions containing suspensions of dispersed nanoparticles can be evaluated using this method [7]. With gamma-ray radiation, such suspensions have been reported to increase the radiolysis yield [1, 2, 11, 14–16].

There are certain drawbacks associated with this method. The system illustrated in Figure 2 is optimally employed with alkaline solutions because of our initial choice of reference electrode (Ag/Ag2O) and the general stability of the Ag
working electrode in an alkaline environment. However, this system has been used to test chloride containing acidic solutions using the Ag/AgCl internal reference. In this case, potential control of the working electrode must be maintained at all times to prevent corrosion and dissolution of the thin Ag layer. This method is also unable to handle nonconductive solutions, such as pure water, due to the basic limitations of an electrochemically based system. The results obtained with 1 mM KOH illustrate this limitation, as the voltage drop resulting from using a solution of low ionic conductivity became significant. For solutions of lower ionic strength/lower conductivity, the voltage drop in solution would have to be further reduced. The voltage drop in solution could be furthered lowered by either (1) moving the counter electrode closer to the working electrode and/or (2) increasing the distance of the flow channel. Both of these options were considered but rejected for the following reasons. Since the working electrode is being used to reduce oxygen or peroxide, the most likely counter electrode reaction is the oxidation of water to produce oxygen. Therefore, it is desirable to keep the counter electrode well separated from the working electrode to prevent oxygen generated at the counter electrode from reaching the working electrode via diffusion. Also, it is desirable to keep the cell volume small, to minimize the amount of solution needed for a measurement, and in the unlikely event of a window failure, to minimize the potential damage to the vacuum system and the accelerator.

Another limitation on solutions to be studied is that, ideally, no species (other than those generated radiolytically) in the solution should be reactive on the Ag electrode at the potential of interest. This limitation provides a low, well-defined background current. In the presence of other electroactive species, it would be necessary to hold the working electrode at the potential of interest for some period of time prior to ion-beam exposure. In the unstirred environment inside the cell, the diffusion limited current for any other electrochemically active species would diminish to a low enough value to allow for subsequent sensing of radiolytically derived reactants, but the sensitivity of the method would be lowered. Lastly, since only O₂ and H₂O₂ are being detected, there is a lack of a closed mass balance. Here, the H₂ yield is simply calculated based on the O₂ and H₂O₂ yields, but these calculations could be verified if H₂ production was independently measured. This could be remedied by circulating the electrolyte and using some means of hydrogen sensing external to the radiolysis cell. However, in this case, the analysis may be considerably more difficult depending on the sensitivity and response time of the hydrogen detection means employed. It should be noted that to the best of the authors’ knowledge, there are no electrocatalysts that are highly active for hydrogen oxidation and that are also inactive for oxygen and/or peroxide reduction. With the appropriate choice of catalyst and potential, a degree of selectivity may be possible, but not the absolute selectivity of silver shown in Figure 3. As a result, an electrochemical means of H₂ detection in the manner employed here for O₂/H₂O₂ is problematic.

These limitations, coupled with the fact that these limitations are not an issue when using a gas chromatograph to analyze the contents of irradiated vials, show that the technique presented here should be complementary to, and not considered a replacement for, the techniques already known in the literature.

5. Conclusions

An electrochemical cell has been developed that allows for the rapid and exhaustive detection of oxygen and/or hydrogen peroxide produced during the radiolysis of aqueous solutions by alpha particle or proton beams. Short, 10–100 s, exposures were sufficient to yield steady-state electrochemical currents proportional to the radiolysis G-factor. The use of thin SiC windows provided a robust means of separating the vacuum environment of the ion-beam accelerator from the aqueous solution at atmospheric pressure with minimal energy loss. The primary advantage of this technique is in its rapid screening capability. Multiple solutions of interest can be analyzed, with repeat measurements for a high level of confidence in the results, in the time it would typically take to make one measurement on one sample using existing techniques. However, given the limitations imposed by the electrochemical detection of the radiolysis products (solutions must be ionically conductive, and the radiolysis products must be able to be selectively oxidized or reduced using a suitable combination of electrocatalyst and potential), this technique provides a complement to, but not a replacement for, the existing analytical methods.

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